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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Anthropogenic influences on the physical state of submicron particulate matter over a tropical forest" *by* Adam P. Bateman et al.

**Anonymous Referee #1**

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**General comments:**

The paper by Bateman describes a results from a field campaign in the Amazon, focusing on the physical state of aerosol particles with an experimental method developed recently by the same group. In particular, particle rebound fraction within an special-purpose impactor setup is used as an indicator of physical state, with the ability to distinguish between liquid particles (i.e., low rebound fraction) and particles of a semi-solid or solid phase state (i.e., medium to high rebound fraction). This method is then used to measure rebound fraction as a function of relative humidity (varied within the impactor; impRH), and ambient parameters such as temperature, humidity, particle O:C ratio, and the concentration of varies gases. It is shown that the particles'



physical state is high at low impRH and low at high impRH, in agreement with previous measurements and with current understanding of the dependence of physical state and of viscosity upon humidity.

What is new in this study is that the particles' physical state varied for periods when the air was influenced by anthropogenic activities such as biomass burning or air originating from the city of Manaus. The higher rebound fraction and, thus, the larger fraction of semi-solid and/or solid particles is in agreement with particle chemistry: the particles are less oxidized and of a lower hygroscopicity than biogenic particles, with a significant contents of C=C double bonds indicative of aromatic VOCs arising from anthropogenic sources. The higher rebound fraction is also in agreement with electron microscopy images showing rather flat particles under conditions of low rebound fraction (and thus impaction of rather low-viscosity liquid) and particles with more pronounced vertical extension und conditions of higher rebound fraction (and thus particles whose shape was likely not influenced during impaction, indicative of more solid-like particles). The authors then utilized aerosol mass spectra as well as various other tracers and parameters measured in parallel to develop a positive-matrix factorization (PMF) analysis that was able to explain the observed rebound fraction to a significant degree.

The manuscript presents a significant contribution to atmospheric aerosol science in the Amazon region and the influence of anthropogenic activities upon the aerosols' physical state. In general, the manuscript was a pleasure to read and the manuscript text and the figures were prepared thoroughly. Moreover, also the measurements appear to have been conducted with great care. In my opinion the manuscript can be published more or less as is, and I have only very minor suggestions that should be taken care of in a revised version.

**Minor and technical comments:**

(1) lines 105-107: '... the anthropogenic molecules have a tendency to reduce water uptake and thereby REDUCE the viscosity of the mixed particles.' I believe you meant to say '... ENHANCE the viscosity'.

(2) lines 101-103: these statements are also supported by the model calculations of Berkemeier et al. (Atmos. Chem. Phys., 14, 12513–12531, 2014.) suggesting that the anthropogenic aromatic SOA precursors naphthalene may lead to higher-viscosity secondary organic material when compared to biogenic precursors such as pinene and isoprene.

(3) In Figure 2b the dashed red line indicating 'pollution at night' is missing. Is this by accident or intentional?

(4) lines 368-379 and Figure 7: the hygroscopicity was measured using different approaches. This is openly discussed in the text, but I would like you to mention that $\kappa_{CCN}$ and $\kappa_{HGF}$ can be quite different for solutes that form non-ideal aqueous solutions. Therefore, I suggest to indicate in the Figure (or at least in the figure caption) that the plotted $\kappa$-values were obtained by different methods at different humidity.

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